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In re Application of Koshiro OCHIAI et al.

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PROCESS FOR PRODUCING POLY(METH)ACRYLATES HAVING
REDUCED METAL CONTENT

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DECLARATION

I, Kazuhiko HASHIMOTO, residing at 9-6-4-106,
Hon-machi, Toyonaka-shi, Osaka, 560-0021, Japan, declare and say:
that I was graduated from Master Course of Graduate School of
Science, Nagoya University in March 1990.

Since April 1990 to the present, I have been employed by
Sumitomo Chemical Co., Ltd., assignee of the above-identified
application, and engaged in research and development in the field of
syntheses of resins for photoresist compositions;

that I am familiar with the invention of the above-identified
application; and

that in order to show the unexpected results obtained by the process for
producing a poly(meth)acrylate of the present invention, I beg to submit the

following experimental data which have been obtained under my supervision:

Experiments

Into 98g of methyl isobutyl ketone were dissolved 74.5g of 2-ethyl-2-adamantyl methacrylate, 35.5g of 3-hydroxy-1-adamantyl methacrylate and 25.5g of α -methacryloyloxy-g-butyrolactone. This was referred to as Monomer solution. Into 33.8g of methyl isobutyl ketone was dissolved 1.97g of 2,2'-azobis(isobutyronitrile). This was referred to as Initiator solution.

Monomer solution and Initiator solution were concurrently added to 206.8g of degassed methyl isobutyl ketone at 87°C and they were allowed to react 6 more hours after concurrent addition. After cooling to room temperature, the reaction mass was obtained.

The reaction mass was then charged to a solution consisting of 2710g of methanol and 677g of ion-exchanged water. After 30 minutes of stirring, the mass was filtrated to obtain solid matter, and the solid matter was charged to a solution consisting of 677g of methanol and 169g of ion-exchanged water. After stirring, the mass was filtrated to obtain solid matter. The charge, stirring and filtration were repeated two more times in the same way as above. Then, it was dried under reduced pressure to give 66.7g of Resin A. A small amount of the dried resin was sampled and the sample was analyzed for various metals in the resin. The results are shown in Table 1.

Example 1

A mixture of 10g of Resin A and 60g of propylene glycol methyl ether acetate was stirred at 40°C until dissolution. Washing with an aqueous oxalic acid consisting of 0.35g of oxalic acid and 17.2g of ion-exchanged water was carried out once at 40°C, followed by phase-separation to obtain organic layer. Then, washing with 17.5g of ion-exchanged water, followed by phase-separation

was repeated 5 times. The obtained organic layer was concentrated to give 30g of Resin solution. Contents of various metals in this solution were measured. The resin concentration in this solution was calculated to be 33% solution from the charged amounts. Values converted to 100% resin are shown in Table 1.

Comparative Example 1 to 3

Experiment was conducted in the same manner as in Example 1 except that the washing was done with an aqueous acetic acid, an aqueous propionic acid or an aqueous malonic acid instead of washing with an aqueous oxalic acid.

Values converted to 100% resin are also shown in Table 1.

Table 1

	Control	Exp.1	Comp.Exp.1	Comp.Exp.2	Comp.Exp.3
Acid	none	Oxalic Acid	Acetic Acid	Propionic Acid	Malonic Acid
Metals	Content (μg/kg)				
Na	150	45	93	510	108
K	25	6	18	24	18
Mg	17	<3	6	33	3
Ca	250	9	21	99	12
Fe	4100	9	33	1200	30
Cu	210	<3	<3	12	<3
Mn	9	<3	<3	<3	<3
Al	68	<3	12	15	6
Zn	1300	<3	51	24	24
Ni	28	<3	<3	9	<3
Cr	23	3	6	9	3
Pb	17	<3	<3	<3	<3

The results in Table 1 show that metal content of the resin obtained in Example 1 is remarkably reduced compared to Comparative Examples 1 to 3.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United State Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 7th day of January 2004.

Kazuhiko Hashimoto
Kazuhiko HASHIMOTO